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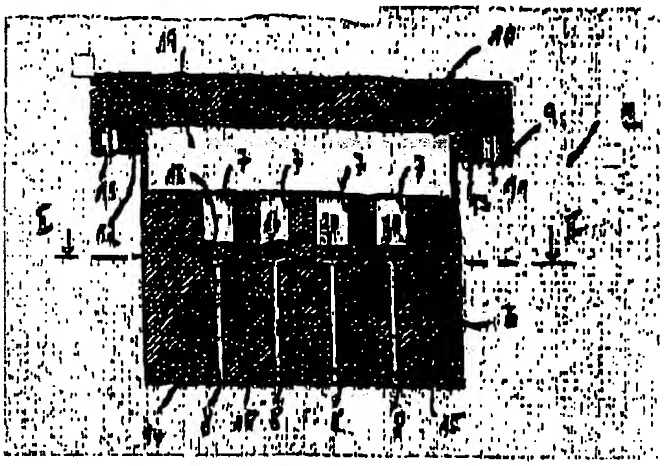
The following data has been taken from the documents that were submitted by the applicant

A request for an examination [of the patent application] has been submitted in accordance with § 44 of the Law on Patents

(54) **Arrangement for testing the catalytic activity of solids that are exposed to a reaction gas**

(57) An arrangement for testing the catalytic activity of solids, which are exposed to a reaction gas, comprises an accommodation unit (1), which has several recesses (7) for accommodating a solid in each case, and a communal supply of gas, and channels (8) that are allotted to the individual recesses so that the solids are all simultaneously exposed to the reaction

gas, and the products, which are formed during the reaction, can be run off separately via the channels and then they can be supplied to an analysis unit. The recesses for accommodating the solids are arranged in the form of a matrix. The test arrangement additi nally comprises a dispensing unit with which the catalysts, which are prepared in a spatially separated manner using combinatorial methods, can be transferred in a simple manner to the recesses of the accommodation unit.



Specification

The invention pertains to an arrangement for testing the catalytic activity of solids, which are exposed to a reaction gas, with a unit for accommodating the solids, and a unit for analyzing the products that are formed during the reaction.

In order to prepare and characterize catalysts, the potentially active carrier components and catalyst components are combined in a supposedly suitable manner, and the multi-component mixtures, which are created in this way, are tested via a suitable test reaction under defined conditions that are usually fixed as a result of technical limitations. Whereas the synthesis of such materials can often be handled with manageable expense, trial runs with the catalysts, by contrast, represent a very time consuming and labor-intensive step. In general, the material that is to be tested is introduced into a laboratory reactor, which has been designed specifically for this purpose, and then, using prescribed parameters, it is tested in terms of its suitability for the reaction of the educt gas mixture in question.

EP 0 423 294 B1 describes a device, which has one single tubular reactor, for investigating and evaluating fluid bed catalysts for cracking purposes. This known device serves for carrying out a series of on-going cyclic experiments while varying the experimental conditions.

The simultaneous testing of catalysts in several separate reactors is also known, whereby these reactors are supplied from a communal gas preparation unit. Supplying the catalysts with the materials, which are to be tested, is also a time consuming and labor intensive procedure when testing catalysts in parallel reactors. The fact that there is a limit, in practice, to the number of reactors that are available for operation in parallel is also a disadvantage.

A decisive innovation in bio-organic syntheses over the last few years has been the development of combinatorial synthetic procedures (Furka, A., Sebastién, E., Asgedom, M., Dibó, G., Abstr. 14th Int. Congr. Biochem., Prague 1988, Vol. 5, 47). In the case of such

syntheses, one usually works with only very small quantities of substances, and use is often made f polymeric carriers for fixing one of the reactants and the products that are formed. Thus, today, combinatorial synthesis represents one of the standard tools in bio-organic chemistry. The efficient testing of a library of substances that has been prepared by such synthetic processes has currently been speeded up most of all in so-called "high throughput screening" for testing potential pharmaceutical agents. Current test methods are based on the addition, from the data bank of substances, of the substance in question to an enzyme or cell cultures that indicate, via luminescence, the presence of an interaction between the substance and a defined active site in the enzyme or in the metabolism of the cell. The substances from the libraries are brought into contact with the test system in question in the form of so-called "arrays", with plates with depressions, which are to be understood to serve as reaction vessels, under the arrays; after this, suitable additional treatment of the sample often takes place as well (irradiation, incubation, etc.), and the concluding evaluation test then follows on from here.

The first attempts to transfer combinatorial synthesis procedures and test procedures to inorganic solids were published by Schultz et al. in 1995 (Xiang, X.-D., Sun, X., Briceno, G., Lou, Y., Wang. K.-A., Chang, H., Wallace-Freedman, W.-G., Chen, S.-W., Schultz, P.G., Science 268, 1995, 1738; and Briceno, G., Chang, H., Sun, X., Schultz, P.G., Xiang, X.-D., Science 270, 1995, 273 as well as Sun, X.-D., Wang, K.-A., Yoo, Y., Wallace-Freedman, W.G., Gao, C., Xiang, X.-D., Schultz, P.G., Adv. Mater. 9, No. 13, 1997, 1046, and Wei, T., Wallace-Freedman, W.G., Schultz, R.G., Xiang, X.-D., Appl. Phys. Lett. 68, 1996, 3506). In this case, use is also made of a coating procedure for the preparation of the compound, whereby this procedure permits various combinations to be prepared on one substrate by means of masking techniques.

The decisive disadvantage of the method that was described by Schultz et al. resides, first of all, in the feature that, as a result of the coating procedure, only a very small proportion of the chemically possible combination[s] is prepared that can be obtained with the wet chemical procedures that are conventional for catalyst preparation. Thus the library of possible catalytically active candidates is considerably reduced in size. A procedure for testing the

catalytic activity of the materials was not described.

A study by Luss et al. has been published in the field pertaining to the simultaneous testing of catalysts (Moates, F.C., Somani, M., Annamalai, J., Richardson, J.T., Luss, D., Willson, R.C., Ind. Eng. Chem. Res., 35, 1996, 4801) in the case of which catalyst pellets were tested by means of infrared thermography. However, this method is limited to reactions with a large evolution of heat. Selectivity differences were not determined.

The problem that forms the basis of the invention is to create an arrangement with which the catalytic activity of solids, which are exposed to a reaction gas, can be tested effectively without high costs in terms of time and labor.

In accordance with the invention, the solution to this problem takes place via the characterizing features of Patent Claim 1.

The arrangement in accordance with the invention permits the simultaneous testing, and also the sequential testing, of a large number of solid catalysts that are exposed to a reaction gas, whereby the conditions such as pressure, temperature, etc. can be varied freely.

The arrangement comprises an accommodation unit that has several recesses for accommodating a solid in each case whose catalytic activity is to be tested. The solids in the accommodation unit are all exposed simultaneously to the reaction gas via a communal gas supply. The stream of gas is run off into the channels that are allotted to the individual recesses so that the products, which are formed via each catalyst, can be analyzed separately for each solid using the analysis unit. This analysis can take place, for example, by means of known spectroscopic, spectrometric or chromatographic procedures. Only one of the channels can, in each case, be freely connected to the analysis unit during the test phase, whereas the gas that is run off via the other channels is not analyzed. However, the accommodation unit also permits the analysis of a gas stream that is run off via several channels. As many channels as is desired can also be closed so that the reaction gas does not flow through them.

In a preferred form of embodiment of the accommodation unit, the recesses are arranged in the form of a matrix. This arrangement in the form of a matrix permits not only the testing of a large number of catalysts in a confined space but, rather, it also facilitates the introduction of the solids, which are to be tested, into the recesses by means of a dispensing unit, in which the catalysts are synthesized, and then they are transferred to the recesses of the accommodation unit. However, it is also possible to synthesize the catalysts directly in the accommodation unit.

The channels for running off the stream of gas are preferably located on the bottom of the recesses so that the reaction gas flows through the catalysts.

The dispensing unit for simultaneously introducing the substances into the accommodation unit has channels that are allotted to the individual recesses, whereby the catalysts can be prepared in a spatially separated manner in these channels by means of combinatorial methods. A sliding valve is advantageously provided in order to be able to close off the channels of the dispensing unit for synthesizing the solids, whereby this sliding valve closes off or, alternately, opens up all the channels.

The channels of the dispensing unit are arranged in a corresponding manner to the recesses of the accommodation unit. The dispensing unit is attached to the accommodation unit in order to transfer the solids, whereby the channels of the dispensing unit are then aligned with the recesses of the accommodation unit.

In order to press the solids out of the recesses, the arrangement in accordance with the invention advantageously comprises a pressing tool that has pressure rams that are allotted to the channels of the dispensing unit.

For adjustment purposes, the accommodation unit has a rotating attachment into which the dispensing unit can be inserted in a well fitting manner. When the dispensing unit has been taken off, a lid is placed on the rotating attachment of the accommodation unit so that a

communal gas supply chamber is created for the recesses in the accommodation unit. The reaction gas can then be supplied to the communal gas supply chamber through a bored out hole in the lid.

In a further preferred form of embodiment, the accommodation unit has a heating device in order to permit adjustment to the desired reaction temperature.

The solid catalysts that are to be tested are preferably positioned on small plates that comprise a porous material, whereby these small plates are arranged in the recesses of the accommodation unit and transversely to the flow direction of the reaction gas.

An example of an embodiment of the arrangement in accordance with the invention for testing the catalytic activity of solids, which are exposed to a reaction gas, will be elucidated in more detail in the following section with reference being made to the drawings.

The following aspects are shown.

Fig. 1 shows, in the form of a sectional illustration, the accommodation unit of the arrangement for testing the catalytic activity of solids that are exposed to a reaction gas,

Fig. 2 shows a section through the accommodation unit along the line II-II of Fig. 1;

Fig. 3 shows the accommodation unit of Fig. 1 in the form of a plan view;

Fig. 4 shows the gas flow diagram for the test arrangement;

Fig. 5 shows the dispensing unit for the test arrangement in the form of a sectional illustration;

Fig. 6 shows a section through the dispensing unit along the line VI-VI of Fig. 5;

Fig. 7 shows the sliding valve of the dispensing unit of Fig. 5 in the form of a plan view;

Fig. 8 shows the pressing tool of the test arrangement in the form of a sectional illustration; and

Fig. 9 shows the size reduction unit of the test arrangement in the form of a sectional illustration.

The arrangement for testing the catalytic activity of solids comprises an accommodation unit 1 (Figs. 1 through 3) in which the solid catalysts are exposed to a reaction gas, an analysis unit 2 together with a valve arrangement 3 (Fig. 4) with which the products, which are formed during the reaction, are analyzed, and a dispensing unit 4 with a pressing tool 5, whereby the catalysts are prepared in a spatially separated manner in the dispensing unit using combinatorial methods (Figs. 5 through 8).

Fig. 1 shows a section through the accommodation unit 1 of the test arrangement. The accommodation unit 1 has a cylindrical element 6 comprising a thermally conductive material, preferably brass, that is provided at its top with cylindrical bored holes 7 that are arranged in the form of a n x m matrix with four rows and four columns (Fig. 2). A 4 x 4 matrix is indicated merely by way of example; arrangements with a significantly larger number of recesses are also possible.

A channel 8 is connected to the bottom of each recess 7, whereby this channel leads out of the accommodation unit 1 at the bottom of the brass element 6. The channels 8 run parallel to one another in the vertical direction. They have a smaller diameter than the recesses. This ensures similar flow rates through all the recesses since the channels represent the main resistances to flow in each case. The individual channels can also be narrowed in a controlled manner in order to permit adjustment to exactly the same flow rates in all the recesses. In order to do this, appropriate throttling devices or similar arrangements can be provided in the accommodation unit.

The inlet of a multi-port valve 24 is connected to the outlet f each channel 8. The multi-port valves are capable of being actuated electromagnetically, and are triggered by a control unit 25 via control lines 26. The first outlet of each multi-port valve 24 is connected to a first communal gas run-off line 28 via a gas line 27, whereby this gas run-off line leads to the analysis unit 2, whereas the second outlet of each multi-port valve 24 is connected to a second communal gas run-off line 30 via a second gas line 29, whereby this communal gas run-off line also leads to the analysis unit 2, e.g. a mass spectrometer.

Depending on the position to which the multi-port valves are switched, the products that are formed by each catalyst during the reaction can be supplied separately and sequentially to the analysis unit 2, or the exiting flow of gas from any two desired catalysts can be analyzed. The valve arrangement, which comprises the multi-port valves, can be expanded at will depending on the number of catalysts that are to be tested.

Instead of a valve arrangement for clearing the individual channels, a manipulator with an inlet capillary, which is capable of being driven in the xyz direction, can also be arranged below the accommodation unit 1, whereby the analysis unit is connected to this inlet capillary. The inlet capillary can then be driven sequentially into the channels by means of the manipulator, so that the exiting gas flow from each catalyst can be analyzed separately. In principle, however, a separate analysis apparatus can also be allotted to each channel, so that analysis with greater parallelism becomes possible.

Fig. 5 shows a section through the dispensing unit 4 of the test arrangement. The dispensing unit 4 has a two-part cylindrical metal element 31 whose diameter corresponds to the internal diameter of the flange 9 of the accommodation unit 1 so that the dispensing unit 4 can be inserted, in a well fitting manner, into the accommodation unit 1. The cylindrical metal element 31 has channels 32 that are arranged in the form of a 4 x 4 matrix. The arrangement and the diameter of the channels 32 correspond to the recesses 7 of the accommodation unit 1, so that these are aligned when the dispensing unit is inserted into the accommodation unit.

A sliding valve 33 is led - in a manner that permits its displacement - between the upper and lower parts 31a, 31b of the cylindrical metal element, and transversely to the longitudinal axis of the channels 32. The sliding valve 33 is a sheet metal plate with bored out holes 34 that are also arranged in the form of a 4 x 4 matrix. These bored out holes 34 are aligned with the channels 32 of the cylindrical metal element 31. The two securing elements 35, which hold the upper and lower parts together, extend through lateral slots 36 in the sliding valve 33, and restrict its sliding path. The sliding valve 33 can be displaced between two positions in which the channels 32 of the dispensing unit are either sealed off or they are opened up.

The sliding valve 33 preferably consists of metal. However, it is also possible for the sliding valve to be a perforated plate comprising a porous material (frit) that serves for filtration when the sliding valve is located in a position in which the bored out holes of the perforated plate are not aligned with the channels of the dispensing unit.

Fig. 6 shows a section through the pressing tool 5 of the test arrangement. The test tool has pressure rams 37 that are arranged in the form of a 4 x 4 matrix, whereby these pressure rams are attached via the screws 38 to a round plate 39. The pressure rams 37 are arranged and designed to have dimensions such that they can be inserted, in a well fitting manner, into the channels 32 of the dispensing unit 4 when the sliding valve 33 is in the open position.

Fig. 7 shows a section through a size reduction unit 40 in the form of a grid that consists of cutting edges 41 that are arranged at right angles at a separation of approximately 2 mm. Prior to inserting the dispensing unit 4 into the accommodation unit, the size reduction unit 40 can be placed in the gas supply chamber 19 of the accommodation unit 1 in order to reduce the size of the solids, which are prepared in the dispensing unit, in order to permit their transfer to the accommodation unit. However, the size reduction unit can also be manufactured in the form of a single integral component along with the dispensing unit.

A description is given in the following section of the oxidation of carbon monoxide on

combinatorially prepared catalysts with use being made of the above test arrangement.

Combinatorially prepared catalysts on the carriers zeolite Y, aluminum oxide, titanium dioxide and zirconium oxide in combination with the potentially catalytically active metals platinum and palladium and the promotor cerium were examined for this test reaction that involves the catalytic oxidation of carbon monoxide to carbon dioxide. In this case, the catalysts were exposed to a reaction gas mixture comprising synthetic air and 80% carbon monoxide. The temperature of the reaction gas atmosphere was increased in steps, and the product gas stream in the individual channels was spectroscopically analyzed for the reaction gas temperature in question.

The catalysts are synthesized in the upper part of the cylindrical element of the dispensing unit in the situation in which the sliding valve is closed. In this case, a different carrier material is deposited in each of the four columns, e.g. a silicate carrier in column 1; aluminum oxide in column 2; titanium dioxide in column 3; and zirconium oxide in column 4. The catalytically active substance is then applied by means of e.g. a fluid impregnation process, e.g. a defined volume of a platinum salt solution in row 1; a defined volume of a palladium salt solution in row 2; a defined volume of a platinum salt solution and a palladium salt solution in row 3; and a defined volume of a platinum salt solution and a palladium salt solution and a cerium salt solution in row 4. The solvent is removed by means of a suitable high temperature treatment. An after-treatment, e.g. with hydrogen, then takes place. In order to be able to transfer the sample array to the accommodation unit, the size reduction unit is placed in the gas supply chamber of the dispensing unit, and the dispensing unit is inserted into the accommodation unit. With the sliding valve in the open position, the solid catalysts are then pressed out of the channels of the dispensing unit by means of the pressing tool, and hence through the grid-like plate and then into the recesses of the accommodation unit. When size reduction of the solids, which have been prepared in the dispensing unit, is not required, placing the size reduction unit in the accommodation unit can also be omitted. The size reduction unit and the dispensing unit are then taken out of the accommodation unit. The accommodation unit is then tightly sealed with the lid, and the reaction gas is supplied. The product gas stream, which is run off from the

catalysts via the individual channels of the accommodation unit, is now analyzed spectroscopically in the analysis unit while the temperature of the reaction gas atmosphere is increased step-wise.

Patent claims

1. Arrangement for testing the catalytic activity of solids, which are exposed to a reaction gas, with a unit for accommodating the solids and a unit for analyzing the products that are formed during the reaction, characterized by the feature that the accommodation unit (1) has several recesses (7) for accommodating a solid in each case, and a communal supply of gas, and channels (8) that are allotted to the individual recesses so that the solids are all simultaneously exposed to the reaction gas, and the products, which are formed during the reaction, can be run off separately via the channels and then they can be supplied to the analysis unit (2).

2. Arrangement in accordance with Claim 1, characterized by the feature that the recesses (7) for accommodating the solids are arranged in the form of a matrix, whereby the channels (8) are connected to the bottom of the recesses (7) so that the reaction gas can flow through the solids that have been introduced into the recesses.

3. Arrangement in accordance with Claim 1 or 2, characterized by the feature that the recesses (7) are cylindrical bored out holes.

4. Arrangement in accordance with one of the Claims 1 through 3, characterized by the feature that the arrangement comprises a dispensing unit (4) that is capable of being attached to the accommodation unit (1) for simultaneously introducing the solids into the accommodation unit.

5. Arrangement in accordance with Claim 4, characterized by the feature that the dispensing unit (4) has channels (32) that are allotted to the recesses (7) of the accommodation unit (1), whereby these solids are synthesized in these channels, and whereby the solids can be transferred

to the recesses of the accommodation unit when the dispensing unit is attached to the accommodation unit.

6. Arrangement in accordance with Claim 5, characterized by the feature that the dispensing unit (4) has a sliding valve (33) that is capable of being displaced between a position for synthesizing the solids, and a position that opens up the channels in order to transfer the solids to the recesses (7) of the accommodation unit (1).

7. Arrangement in accordance with Claim 6, characterized by the feature that the sliding valve (33) is a perforated plate comprising metal or a porous material.

8. Arrangement in accordance with one of the Claims 1 through 7, characterized by the feature that the accommodation unit (1) has a rotating attachment (9) into which the dispensing unit (4) is capable of being inserted, and a lid (10), which is capable of being positioned on the rotating attachment (9) to form a communal gas supply chamber when the dispensing unit has been taken off, whereby the lid is provided with at least one bored out hole (17) for supplying the reaction gas.

9. Arrangement in accordance with one of the Claims 4 through 8, characterized by the feature that the arrangement comprises a pressing tool (5) with pressure rams (37) that are allotted to the channels (32) of the dispensing unit (4), whereby these pressure rams can be introduced into the channels (32) of the dispensing unit in order to press out the solids.

10. Arrangement in accordance with one of the Claims 1 through 9, characterized by the feature that the accommodation unit has a heating device (24).

11. Arrangement in accordance with one of the Claims 1 through 10, characterized by the feature that small plates (20), which are made from a porous material, are provided in the recesses (7) of the accommodation unit (1), whereby these small plates are provided for the accommodation f the solids that are to be introduced into the recesses.

12. Arrangement in accordance with one of the Claims 4 through 11, characterized by the feature that the arrangement has a siz reduction unit (40) for reducing the particle size of the solids that are prepared in the dispensing unit (4) in order to permit their transfer to the accommodation unit (1).

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3 page(s) of drawings attached hereto

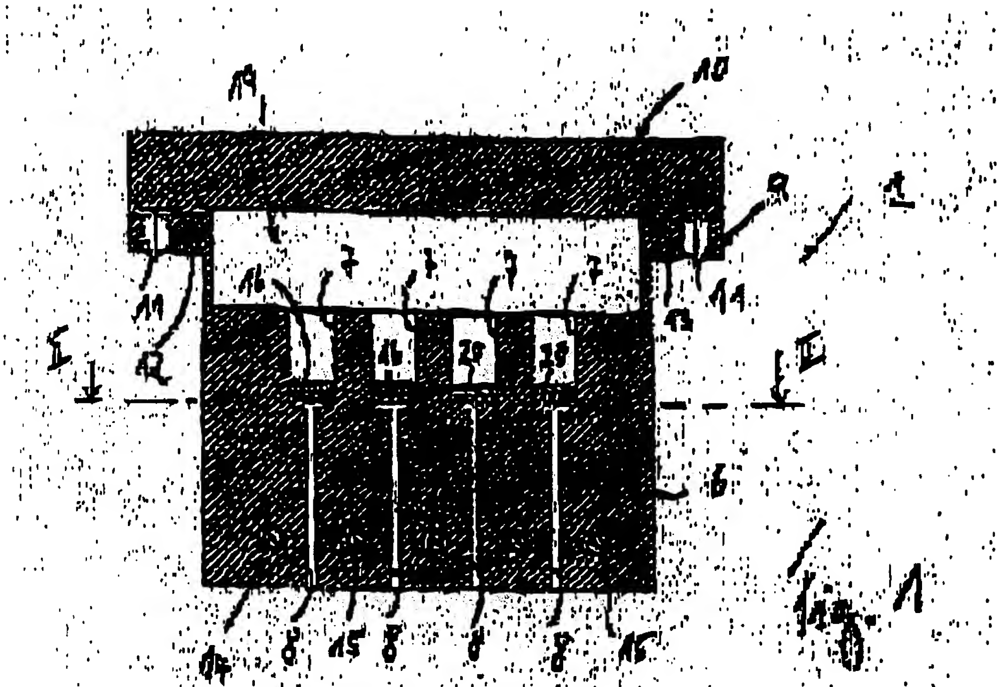

fig. 1

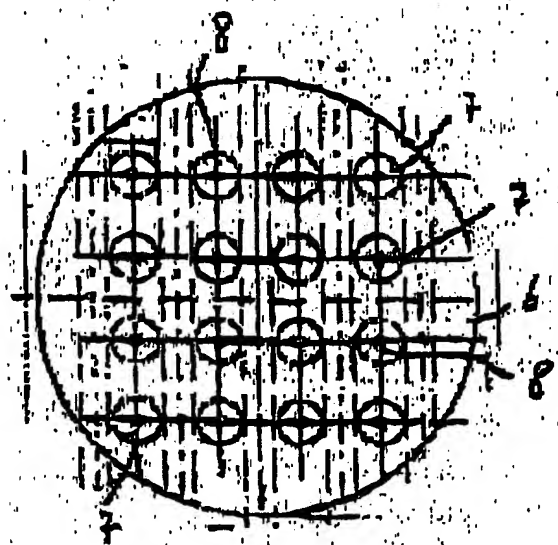

fig. 2

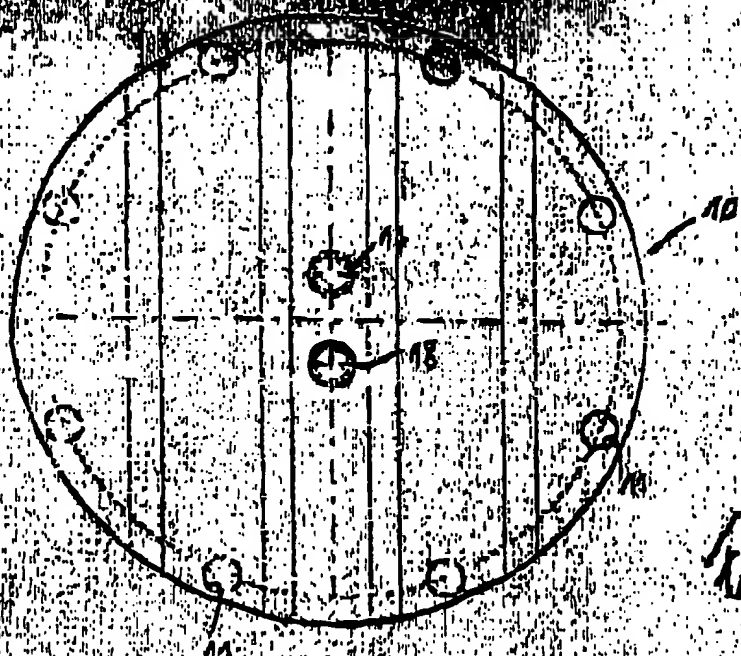

Fig. 3

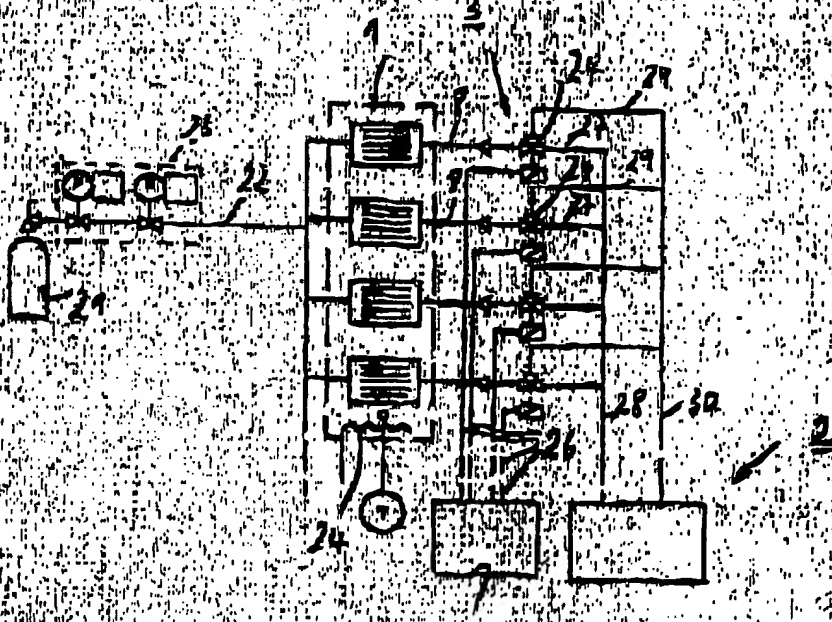

Fig. 4

Fig. 8

Fig. 9

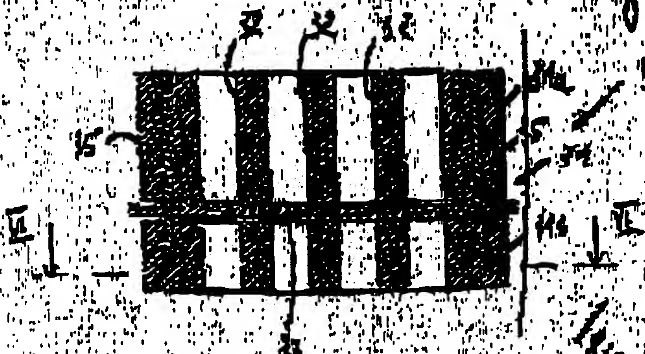

Fig. 5

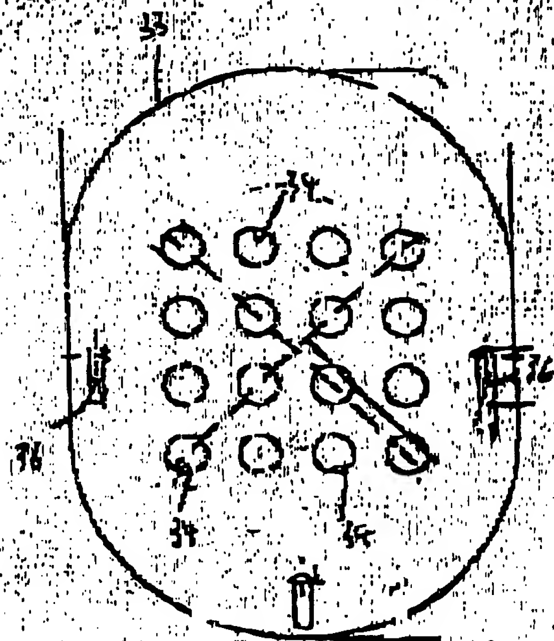

Fig. 7

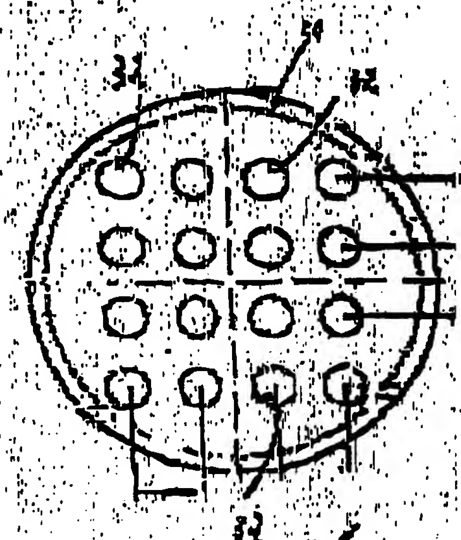

Fig. 6